THz generation by optical rectification and competition with other nonlinear processes

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Z.Y. Zhao, S. Hameau, M. Voos, and J. Tignon

***Abstract*—We present a study of the terahertz (THz) generation by optical rectification in a 2 mm thick <110> ZnTe crystal. The conditions for the most efficient THz generation are described. We investigate the competition between optical rectification (OR), second harmonic generation (SHG), two-photon absorption (TPA) and free-carrier absorption in the diffraction limit, for excitation spot sizes smaller than the THz wavelength. We find that while free-carrier absorption contributes significantly to reduce the THz emission, this contribution does not depend on the excitation spot size. On the contrary, two-photon absorption induces a strong dependence of the THz emitted power for decreasing excitation sizes.**

***Index Terms*—Terahertz, optical rectification, two-photon absorption, second harmonic generation, free-carrier absorption.**

I. Introduction

ULTRA-FAST THz spectroscopy is a powerful technique for studying a wide variety of materials including gases, liquids and solids [1]. In this context, a now widely used technique consists in generating THz pulses by optical rectification of femtosecond laser pulses [2,3]. An ultra-fast optical pulse is focused on a nonlinear crystal, which then radiates a THz pulse with a duration of few cycles of the electromagnetic field. In the spectral domain, the radiation is broadband, from about 100 GHz to typically 3 THz for a 100 fs pulse. This process can also be seen as the result of the frequency difference between all the frequency components present within the optical pulse. For an excitation with a typical Ti:sapphire mode-locked laser, the generated THz power is in the range of only a few tens of nW, which stresses the importance of understanding and optimizing the THz generation in nonlinear crystals.

Efficient optical rectification requires using materials with large second-order nonlinear susceptibilities and well-suited phase-matching properties for THz generation. ZnTe crystals with a <110> orientation offer one of the best compromises in this context [4]. Unfortunately, when a ZnTe crystal is irradiated with a high-power laser pulse, other competing nonlinear processes such as second harmonic generation, two-photon absorption and subsequent free-carrier absorption may also occur, resulting in a decrease of the THz generation [5,6,7]. Additional competing mechanisms have also been put forward, such as the fact that upon tight focusing, the size of the THz source (the laser spot size) becomes smaller than the typical THz wavelength, resulting in a decrease of the THz generation due to diffraction [8]. Nevertheless, to date, existing analysis of this competition are not comprehensive and the explanations are sometimes even contradictory.

In the present work, we investigate the THz generation by optical rectification in a <110> 10x10x2 mm ZnTe crystal as a function of power, focusing and crystal orientation. The conditions for the most efficient THz generation are described. Upon tight focusing, for excitation spot sizes smaller that the THz wavelength, SHG, TPA as well as free-carrier absorption are observed while the THz emission is strongly reduced. We find that free-carrier absorption cannot be neglected as often assumed in the past. Nevertheless, because of diffraction effects, this contribution is excitation-size independent upon tight focusing. On the contrary, TPA, which also contributes to the reduction of the THz emission, strongly depends on the excitation-spot size.

II. Experimental results

A. Optical rectification and THz emission

The excitation of the <110> 2 mm-tick ZnTe crystal is provided by a 800 nm Ti: sapphire laser (100 fs pulses) at 300 mW, using a 4 cm focusing lens. The ZnTe crystal is virtually transparent in the frequency domain of interest (0.1 – 3 THz). A liquid helium cooled bolometer connected to a lock-in amplifier was used to detect the spectrally integrated THz emission (after proper filtering of the transmitted 800 nm laser beam).

Figure 1 shows the THz emission as a function of the ZnTe azimuthal angle (closed symbols). The THz intensity is proportional to the square of the nonlinear polarization which angle dependence (angle between the polarization of the optical pump beam and the [001] axis) is determined by the nonlinear susceptibility tensor [9]. Figure 1 also displays the angle dependence of the measured SHG (open symbols) generated in the ZnTe crystal upon focusing (z = 0, where z is the distance between the beam waist and the crystal), which shows maxima and minima for the same azimuthal angles.

Manuscript received May 11, 2006.

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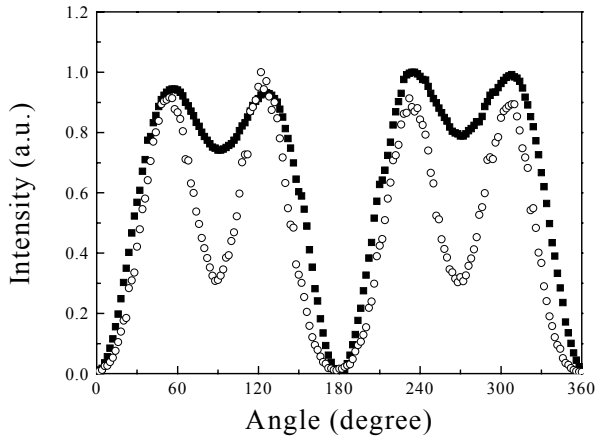

Fig. 1. Closed symbols : THz intensity (bolometric detection) as a function of the azimuthal angle. Open symbols : SHG intensity (measured with a PMT) upon focusing (z = 0), as a function of the azimuthal angle. Excitation power 300 mW.

Figure 2 shows the angle dependence of the THz emission (right axis) as well as the angle dependence of the direct transmission at 800 nm (measured with a simple photodiode) when the laser beam is focused (z = 0). Under proper crystal angle orientation, the transmission at 800 nm is reduced by about 10 % and a yellow-green luminescence induced by TPA can be observed on the ZnTe crystal surface. In principle, optical rectification, SHG as well as TPA can be responsible for the optical pump depletion for specific azimuthal angles. Nevertheless, the optical rectification efficiency is only on the order of 10^{-7} and, as already pointed out in previous studies [7], when rotating the ZnTe crystal, the SHG generation changes by several orders of magnitude whereas the transmission at 800 nm only varies by about 10 %. Thus, optical rectification and SHG are not responsible for the optical pump depletion.

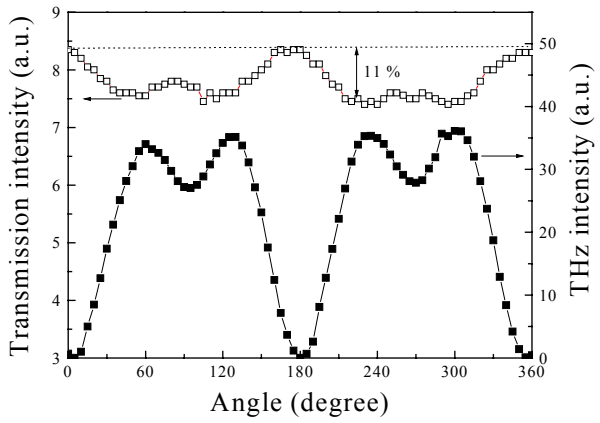

Fig. 2. Closed symbols : THz intensity (bolometric detection) as a function of the azimuthal angle. Open symbols : transmission at 800 nm (measured with a photodiode) upon focusing (z = 0), as a function of the azimuthal

Easy crystal axis determination is highly desirable when developing ultra-fast THz spectroscopy experiments based on optical rectification and Free-Space-Electro-Optical-Sampling (which relies on a second ZnTe crystal). In this case, several parameters need to be properly adjusted simultaneously (ZnTe crystals orientation, optical pump focusing, time delay and spatial overlap between the THz pulse and the reference pulse, etc.), which renders the alignment tedious. In this context, Figure 2 also shows that the angle-dependence of the transmission is the simplest method for determining the crystal orientation (with the exception of very thin crystals).

B. Z-scan experiment

Competition between optical rectification and other nonlinearities was investigated using a modified z-scan method [7]. The THz emission, the SHG emission as well as direct transmission at 800 nm were measured as a function of the distance *z* between the beam waist and the crystal, along the optical axis for an optical pump power of 300 mW. The waist radius decreases down to about 10 µm upon focusing (z = 0), corresponding to a Rayleigh length of about 0.4 mm. As a consequence, in the following, the laser spot radius w(z) lies mostly in the asymptotic region where it can be well approximated by $w(z) \propto z$. For small distances z (tight focusing), SHG generated by the ZnTe crystal is measured using a photomultiplier (after filtering the direct transmission at 800 nm).

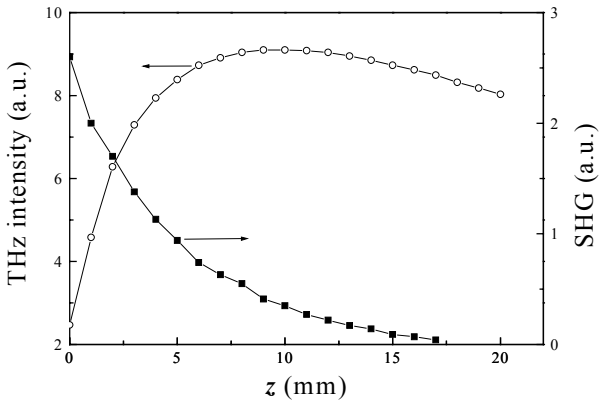

Fig. 3. Open symbols : THz intensity (bolometric detection) as a function of the distance to focus point. Open symbols : Second Harmonic Generation as a function of the distance to focus point. Focal 4 cm. Power 300 mW.

Figure 3 shows the THz emission measured as a function of the distance z to the focus (left axis) as well as the SHG (right axis). At first sight, the easiest method to increase the THz emission efficiency is to focus the optical pump beam on the nonlinear ZnTe crystal. Indeed, for large z, the THz emission first increases when decreasing the laser spot size. But the emission then tends to saturate for z on the order of 10 mm. For even smaller distances, the THz emission drops significantly, forming a 5 mm large 'z-hole'. Hence, as often reported, the best THz emission efficiency is obtained when slightly defocusing the optical pump from the ZnTe crystal. Several mechanisms have been put forward to explain the formation of this 'z-hole', among which optical pump depletion by TPA, THz absorption by free carrier absorption, or diffraction effects when the laser spot radius becomes comparable with the THz wavelength (which

occurs here when *z* is about 10 mm).

C. Two-color excitation

The free-carrier absorption was estimated using a two-color experiment. The free-carrier absorption is proportional to the carrier density in the ZnTe crystal generated at the second harmonic energy. The 800 nm laser beam is then transmitted through a BBO crystal prior to be focused on the ZnTe crystal. For this experiment, the distance z between the 800 nm beam waist and the ZnTe crystal is adjusted in order to maximize the THz emission while minimizing the SHG generated in the ZnTe crystal (z = *15 mm*). In a first measurement, the BBO is not phase-matched (BBO angle adjusted to obtain zero SHG). We then measure the THz emission in the absence of any competing nonlinearities within the ZnTe crystal. In a second measurement, the BBO is phase-matched to generated SHG in order to provide a two-color excitation of the ZnTe crystal. Here, the azimuthal angle of the BBO crystal is adjusted in order to obtain a SHG on the order of the SHG emission observed when exciting the ZnTe crystal at z = *0*. The measured THz emission is strongly reduced, corresponding to an absorption coefficient α_{THz} of about 4 cm^{-1}, showing that free-carrier absorption of the THz radiation cannot be neglected in this problem, contrary to what was previously assumed [6,7,8].

III. INTERPRETATION

A. Diffraction limit

As already pointed out above, the phenomena discussed here occur for *z* distances much larger the optical beam Rayleigh length and the laser spot radius w(z) can well be approximated by its asymptotic limit ($w(z)=\lambda z/\pi w_0$, where w_0 is the beam waist. The spot size $S \propto z^2$). Since optical rectification is a second-order nonlinear process, the THz electric field E_{THz} is proportional to the laser intensity $I_0 = P_0 / S$, where P_0 is the input power. When diffraction effects can be neglected (spot size larger than the THz wavelength), the measured THz signal power is thus $P_{signal} \propto S\,(P_0 / S)^2 \propto S^{-1} \propto z^{-2}$. This z^{-2} dependence of the THz signal is indeed observed for large distances (z > 13 mm) as can be seen in Figure 4 (dashed line). On the contrary, as pointed out by Dakovski *et al*. [6], the situation is different when the THz emission is generated by a localized emitter of extension smaller than the THz wavelength (centered close to 300 μm). This typically occurs for z smaller than 10 mm. In this case, the THz emission from the whole emitter section interfere constructively and the THz emitted power now reads : $P_{signal} \propto (S\,E_{THz})^2 \propto (S\,(P_0 / S))^2 \propto S^0 \propto z^0$. As a consequence, the THz emitted power is expected to become independent of the spot size close to focus. This is sketched by the dotted line in Figure 4.

B. Free-carrier absorption

The equation describing the generation of THz has the form:

$$\frac{dE_{THz}}{dz} = aI_0 - \alpha_{THz} E_{THz}$$

where the first term corresponds to the generation of THz by optical rectification and the second term to the THz absorption by free carriers. In this situation, $E_{THz} \propto \eta\, a\, l\, I_0$, where l is the crystal length and $\eta = \left(1 - e^{-\alpha_{THz} l}\right) / \alpha_{THz}\, l$ is a factor that reduces the THz field induced by optical rectification. As a consequence, in the diffraction limit (since z is small in this situation), the THz power signal is now $P_{signal} \propto (S\,E_{THz})^2 \propto (S\,(\eta P_0 / S))^2 \propto S^0 \propto z^0$. The THz emission is globally reduced by a factor η^2, which can be very large, but still independent on the spot size. This is sketched in Figure 4 (dot-dashed line).

C. Two-photon absorption

As already discussed above, two-photon absorption is responsible for the depletion of the optical pump beam. The equation for the nonlinear absorption is:

$$\frac{dI}{dz} = -\beta I^2$$

in which the linear absorption at the optical frequency is neglected and where β is the nonlinear absorption coefficient. This results in the following optical intensity in the crystal:

$$I(z) = \frac{I_0}{1 + \beta I_0 z}$$

In this case, the emitted THz signal in the diffraction limit becomes:

$$P_{signal} \propto P_0^2 \Big/ \left(1 + \beta P_0 l \frac{\pi w_0^2}{\lambda^2 z^2}\right)^2$$

which strongly depends on z and creates a z-hole as already mentioned in Ref. [6]. In the presence of free carrier

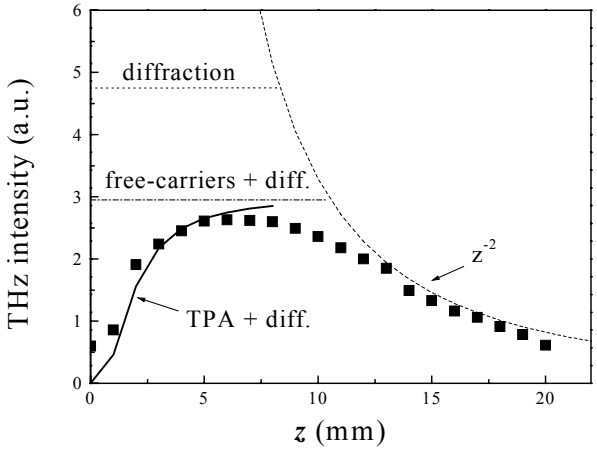

Fig. 4. Symbols : measured THz emission as a function of the distance to focus. Long dashed line : Theoretical THz emitted power beyond the diffraction limit. The THz emission decreases quadratically with the spot size and with the distance to focus. Dotted line: Theoretical THz emitted power in the diffraction limit (no competing nonlinearities included). The THz emission is then independent of the excitation spot size. Dot-dashed line: Theoretical THz emission in the diffraction limit when free-carrier absorption is included. The THz emission remains independent of the excitation spot size. Solid line: Theoretical THz emission in the diffraction

absorption, the input optical power in the above expression is multiplied by a factor η.

Fitting the z-hole in Figure 4 leads to a nonlinear absorption coefficient β on the order of 10 cm/GW, in good agreement with values reported in the literature [6,7,10].

Last, we remark that the above equation does not hold for $z \approx 0$ because the asymptotic approximation for the spot size radius does not hold any more. In this approximation, the spot size is zero for $z = 0$ and the fitted THz signal then drops exactly to zero at the focus. Taking into account the Gaussian profile of the beam leads to a finite THz emission at $z = 0$, as in the experiment.

IV. DISCUSSION

Diffraction effects are essential in the understanding of the THz emission when the pump beam is focused. As shown by Gaivoronskii *et al*. [7], if diffraction effects are not taken into account, TPA reduces the optical pump power, and thus the THz emission, but without forming a z-hole. In this case, TPA and SHG cannot explain this effect even qualitatively. On the contrary, when diffraction effects are properly taken into account, TPA can be shown to result in the formation of a strong reduction of the THz emission upon focusing in the form of a z-hole. Here, we also show that free-carrier absorption cannot be neglected in this analysis since it is responsible for a strong absorption of the THz radiation. Nevertheless, free-carrier absorption only tends to scale down the measured THz power and cannot, by itself, explain the formation of a z-hole.

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